

Search for long-lived isomeric states in neutron-deficient thorium isotopes

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The discovery of naturally occurring long-lived isomeric states ($t_{1/2} > 10^8$ yr) in the neutron-deficient isotopes $^{211,213,217,218}\text{Th}$ [A. Marinov *et al.*, Phys. Rev. C **76**, 021303(R) (2007)] was reexamined using accelerator mass spectrometry (AMS). Because AMS does not suffer from molecular isobaric background in the detection system, it is an extremely sensitive technique. Despite our up to two orders of magnitude higher sensitivity we cannot confirm the discoveries of neutron-deficient thorium isotopes and provide upper limits for their abundances.

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I. INTRODUCTION

Recently the discovery of “high spin super- and hyperdeformed” isomeric states in neutron-deficient thorium isotopes was claimed [1]. The authors investigated solutions of natural thorium and announced the discovery of long-lived isomers in $^{211,213,217,218}\text{Th}$. They conclude that the lower limit of the half-lives must be 10^8 years and assume that these might belong to “high spin K -type isomers,” which were “preferentially produced by heavy ion reactions.”

These observations are extremely surprising for several reasons. The ground states of the thorium isotopes in question are all α emitters with half-lives ranging from 144 ms down to 117 ns for the $N = 128$ nucleus ^{218}Th , which has one of the shortest half-lives ever measured for a nuclear ground state. In Table I we list the available information on these thorium isotopes. Even if the electromagnetic decay for the isomers claimed by Ref. [1] would be so slow due to K hindrance, in addition the α decay would have to be slowed down because of angular momentum hindrance by at least 16 to 22 orders of magnitude to explain the long half-lives required for their natural occurrence.

Furthermore there is no way within the current understanding of nucleosynthesis these states could have been produced before the formation of the solar system.

The stellar nucleosynthesis of isotopes heavier than iron occurs predominantly via neutron capture reactions in equal shares in the so-called slow and rapid processes (“ s process” and “ r process”) [8]. The neutron density in the s process is in the range $N_n \approx 10^8 \text{ cm}^{-3}$, forcing the reaction path to follow the line of β stability. The s -process nucleosynthesis terminates in the reaction cycle $^{209}\text{Bi}(n, \gamma)^{210}\text{Bi}(\beta^-)^{210}\text{Po}(\alpha)^{206}\text{Pb}$. To populate thorium isotopes in the s process, a bridge of nuclear states with half-lives in the order of years between ^{209}Bi and these thorium isotopes would be necessary.

The long-lived primordial actinides ^{232}Th , ^{235}U , and ^{238}U are produced in the high neutron density scenario ($N_n \gg 10^{20} \text{ cm}^{-3}$) of the r process. The exact stellar production site is still under discussion, the most probable scenarios are

core-collapse supernovas or neutron star mergers. The starting seed for both the r and s processes is iron peak nuclei produced by previous fusion processes (“burning stages”) in the star. Because of its large neutron density the r -process reaction path is driven far away from stability close to the region of the neutron drip line.

By this mechanism the r process is able to climb within a few hundred milliseconds up to its termination point around $A \approx 260$ ($Z = 94$, Pu), where “fission recycling” by spontaneous and β -delayed fission sets in and transfers some material back into the mass region $A \approx 130$. When the neutron production ends and/or the temperature decreases (“freeze-out”) the short-lived radioactive nuclei decay back to stability via long β -decay chains. This produces the well-known observed solar r -abundance peaks at $A \approx 80$, 130, and 195 [9], corresponding to the very neutron-rich r -process progenitor nuclei with $N = 50$, 82, and 126.

Beyond $A = 210$ these long decay chains will reach α emitters that are (almost) stable against β decays. The transmutation of these radioactive isotopes will then proceed via α -decay chains, ending either in bismuth and the lead isotopes or in the long-lived actinides ^{232}Th , ^{234}U , ^{235}U , and ^{238}U . In any case the nuclides finally populated in the r process have a considerably higher A/Z ratio than the values $A/Z \leq 218/90$ for the thorium isotopes in question.

Thus the findings of Ref. [1] motivated us to repeat the measurements with an independent method.

II. EXPERIMENTAL TECHNIQUE

Accelerator mass spectrometry (AMS) is a highly sensitive method to determine small traces of radionuclides without interfering background from molecules. For the measurement we used our AMS setup at the Maier-Leibnitz-Laboratory in Garching (see Fig. 1).

The thorium sample was pressed in a graphite cathode for the sputter ion source from where ThC^- molecules of different masses were extracted. The sample was prepared by mixing ThO_2 (Merck LotNr. 2353854) with graphite powder.

The first mass selection of the molecules was performed with the 90° injector magnet, which has a mass resolution of $1/200$. For an additional energy selection on the low energy side we used an 18° electrostatic deflection.

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TABLE I. Decay properties of $^{211,213,217,218}\text{Th}$ and known isomers, with excitation energy, isospin, and parity.

Isotope	I^π	E_{ex} (keV)	$t_{1/2}$	Ref.
^{211}Th	?	0	(40^{+3}_{-1}) ms	[2]
^{213}Th	$(5/2)^-$	0	(144 ± 21) ms	[3]
$^{213}\text{Th}^m$	$(13/2)^+$	1180	(1.4 ± 0.4) μs	[4]
^{217}Th	$(9/2^+)$	0	(241 ± 5) μs	[5]
$^{217}\text{Th}^m$	$(25/2^+)$	“2252+X”	(67^{+17}_{-11}) μs	[6]
^{218}Th	0^+	0	(117 ± 9) ns	[7]

The terminal voltage of the MP tandem accelerator was set at voltages around 11 MV and thus the negatively charged molecules are accelerated to an energy of about 11 MeV. The thin carbon stripper foil ($4 \mu\text{g}/\text{cm}^2$) in the high voltage terminal destroys these molecules completely. Positive ions in high charge states are produced and accelerated further on the high energy side. The thorium ions had typically an energy of around 120 MeV. The beam transport system was adjusted with $^{209}\text{Bi}^{10+}$ ions from $^{209}\text{BiO}^-$. This setting was verified with $^{232}\text{Th}^{10+}$ from $^{232}\text{ThC}^-$.

On the high energy side of the accelerator the charge state 10^+ was selected by a Wien velocity filter and a high resolution 90° analyzing magnet. To transport different ions, all the magnetic elements after the accelerator were untouched and only the terminal voltage and the voltage of the Wien filter were adjusted. The parameters of the beam transport for the neutron-deficient thorium isotopes were calculated with the masses given in Ref. [1]. The acceptance of our beam transport system has a value of about $\Delta m/m \approx \pm 4 \cdot 10^{-4}$.

The ions are identified by a time-of-flight spectrometer, where the start signal is delivered by a microchannelplate

detector. An ionization chamber determines the energy loss of the ions and a silicon surface barrier counter measures the residual energy and acts as the stop detector simultaneously. The setup of the detection system is described in more detail in Ref. [10].

From time to time a beam of ^{232}Th , attenuated by a factor of 10^6 with two low transmission grids, is sent to the detector for normalization of the flux of the thorium isotopes. The count rate of the unattenuated beam of ^{232}Th corresponds to about 10^8 Hz for the various measurements. The number of detected events of the less abundant thorium isotopes is normalized to this ^{232}Th particle current. Scattering inside the attenuator grids and thus loss of transmission were taken into account by the measurement of the sum of the daughter products ^{228}Th and ^{228}Ra . They cannot be distinguished by the described detection method. As RaC^- and ThC^- have a different formation rate in a cesium sputter ion source, the measured $A = 228$ particle flux is corrected according to the data in Refs. [11] and [12]. The employed ThO_2 is more than 15 years old, so ^{228}Ra and ^{228}Th should be in equilibrium. Thus the transmission of the different thorium isotopes of interest versus the attenuated ^{232}Th is accounted for by multiplying the ^{232}Th yield with a factor of 1.2 to 1.4. The run time for each isotope lasted several hours.

III. RESULTS

None of the four neutron-deficient thorium isotopes could be detected with an abundance of 1 to $10 \cdot 10^{-11}$ as suggested in Ref. [1].

For the two nuclides ^{211}Th and ^{217}Th , there were no events in the expected regions of the time-of-flight signal and the energy signal. Background events were detected at a wide range of different time-of-flights and energies due to scattered ions for ^{232}Th in the 11^+ or 10^+ charge state. One event was measured in the expected range for ^{213}Th and ^{218}Th , respectively, which would correspond to an abundance of these isotopes on the level of several 10^{-13} . But these single events cannot be distinguished from the outspread background.

Upper limits for the yield were estimated from the measurement values of run times, particle flux, and transmission, taking into account 1σ errors of these quantities as a consequence of several uncertainties. Furthermore, statistical analysis of small signals according to Ref. [13] was included.

The value for the additional extraction of ^{228}Ra has an uncertainty of 10% [11]. The uncertainty of the attenuation is assumed to be 5%. The particle currents of mass 228 and mass 232, used to determine the transmission and the flux of the thorium ions are subject to a statistical fluctuation of 5%.

Our measurements contradict the observations of Marinov *et al.* [1]. The resulting upper limits (see Table II) are about one order of magnitude below the values given in Ref. [1] and are in accordance with results from Dellinger *et al.* [14].

IV. DISCUSSION

The values of our AMS measurements in thorium samples demonstrate that in nature no neutron-deficient isotopes

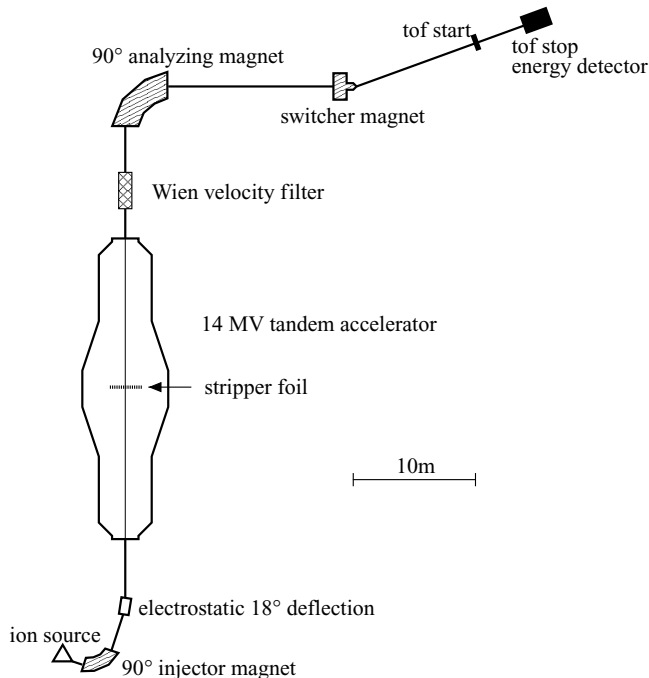


FIG. 1. AMS setup for the detection of $^{211,213,217,218}\text{Th}$.

TABLE II. AMS results for the abundance of $^{211,213,217,218}\text{Th}$, upper limits given at the 68.3% confidence level.

Isotope	Events	Upper limit for $A\text{Th}/^{232}\text{Th}$
^{211}Th	0	$9.6 \cdot 10^{-13}$
^{213}Th	1	$1.2 \cdot 10^{-12}$
^{217}Th	0	$6.6 \cdot 10^{-13}$
^{218}Th	1	$2.4 \cdot 10^{-12}$

of thorium at masses 211, 213, 217, and 218 exist with abundances of 1 to $10 \cdot 10^{-11}$ relative to ^{232}Th . Measurements were stopped as soon as it became clear that the result of the concentration of neutron-deficient thorium isotopes in our sample is about an order of magnitude below the values from Ref. [1]. So our sensitivity is limited by the run time more than by the background because of scattered ^{232}Th ions.

There is also no scenario of nucleosynthesis besides the r process that is able to produce isotopes beyond ^{209}Bi in such amounts that they would be somehow detectable on earth. However, the r process cannot reach neutron-deficient thorium isotopes like $^{211,213,217,218}\text{Th}$ in any way because they are located in the midst of pure α emitters. The lightest thorium isotope that can be reached by the r -process β -decay chains is ^{226}Th .

The argument to attribute these long-lived isomers to K isomers [15] can also be disproved. Most K isomers were identified around $A \approx 180$ (e.g., $^{180}\text{Ta}^m$) where large mid-shell deformations with neutrons and protons occupying high-lying orbitals can be found. The heaviest K isomers found up to now are in ^{250}Fm [16], ^{256}Fm [17], ^{252}No [18], ^{254}No [16,19], and ^{270}Ds [20]. These isomeric states have half-lives in the order of μs to ms. Prolate deformations are an important condition for the existence of K isomers. This requirement is not fulfilled for the neutron-deficient thorium isotopes. The aforementioned superheavy nuclei exhibit ground-state quadrupole deformations between $\beta_2 = 0.221$ and 0.246 (prolate), whereas for ^{211}Th the maximum deformation is calculated to be $\beta_2 = -0.105$ (oblate deformation) [21]. Also ^{213}Th and ^{217}Th are estimated to be slightly oblate, whereas the even-even nucleus ^{218}Th , two neutrons away from the shell closure at $N = 126$, is expected to be almost spherical ($\beta_2 = 0.008$). Thus no K isomers should exist in $^{211,213,217,218}\text{Th}$.

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- [1] A. Marinov, I. Rodushkin, Y. Kashiv, L. Halicz, I. Segal, A. Pape, R. V. Gentry, H. W. Miller, D. Kolb, and R. Brandt, Phys. Rev. C **76**, 021303(R) (2007).
- [2] E. Browne, Nucl. Data Sheets **103**, 183 (2004).
- [3] M. Basunia, Nucl. Data Sheets **108**, 633 (2007).
- [4] J. Khuyagbaatar, S. Hofmann, F. P. Heßberger *et al.*, Eur. Phys. J. A **34**, 355 (2007).
- [5] Y. Akaoli, Nucl. Data Sheets **100**, 141 (2003).
- [6] P. Kuusiniemi, F. P. Heßberger, D. Ackermann, S. Hofmann, B. Sulignano, I. Kojouharov, and R. Mann, Eur. Phys. J. A **25**, 397 (2005).
- [7] A. Jain and B. Singh, Nucl. Data Sheets **107**, 1027 (2006).
- [8] E. M. Burbidge, G. Burbidge, W. A. Fowler, and F. Hoyle, Rev. Mod. Phys. **29**, 547 (1957).
- [9] E. Anders and N. Grevesse, Geochim. Cosmochim. Acta **53**, 1997 (1989).
- [10] C. Wallner, T. Faestermann, U. Gerstmann, W. Hillebrandt, K. Knie, G. Korschinek, C. Lierse, C. Pomar, and G. Rugel, Nucl. Instrum. Methods Phys. B **172**, 333 (2000).
- [11] X.-L. Zhao, M.-G. Nadeau, M. A. Garwan, L. R. Kilus, and A. E. Litherland, Nucl. Instrum. Methods Phys. B **92**, 258 (1994).
- [12] S. G. Tims, G. J. Hancock, L. Wacker, and L. K. Fifield, Nucl. Instrum. Methods Phys. B **223**, 796 (2007).
- [13] G. J. Feldman and R. D. Cousins, Phys. Rev. D **57**, 3873 (1998).
- [14] F. Dellinger, O. Forstner, R. Golser, W. Kutschera, A. Priller, P. Steier, A. Wallner, and G. Winkler, Poster presented at AMS 11, Rome, 2008.
- [15] P. Walker and G. Dracoulis, Nature **399**, 35 (1999).
- [16] A. Ghiorso, K. Eskola, P. Eskola, and M. Nurmi, Phys. Rev. C **7**, 2032 (1973).
- [17] H. L. Hall, K. E. Gregorich, R. A. Henderson, D. M. Lee, D. C. Hoffman, M. E. Bunker, M. M. Fowler, P. Lysaght, J. W. Starnes, and J. B. Wilhelmy, Phys. Rev. C **39**, 1866 (1989).
- [18] B. Sulignano, S. Heinz, F. P. Heßberger, S. Hofmann *et al.*, Eur. Phys. J. A **33**, 327 (2007).
- [19] S. K. Tandel, T. L. Khoo, D. Sewerniak, G. Mukerjee *et al.*, Phys. Rev. Lett. **97**, 082502 (2006).
- [20] S. Hofmann, F. P. Heßberger *et al.*, Eur. Phys. J. A **10**, 5 (2001).
- [21] P. Möller, J. R. Nix, W. D. Myers, and W. J. Swiatecki, At. Data Nucl. Data Tables **59**, 185 (1995).